

Development of a mechanistic source term approach for liquid-fueled Molten Salt Reactors

Chemical & Fuel Cycle Technologies Division

About Argonne National Laboratory

Argonne is a U.S. Department of Energy laboratory managed by UChicago Argonne, LLC under contract DE-AC02-06CH11357. The Laboratory's main facility is outside Chicago, at 9700 South Cass Avenue, Argonne, Illinois 60439. For information about Argonne and its pioneering science and technology programs, see www.anl.gov.

DOCUMENT AVAILABILITY

Online Access: U.S. Department of Energy (DOE) reports produced after 1991 and a growing number of pre-1991 documents are available free at OSTI.GOV (<http://www.osti.gov/>), a service of the US Dept. of Energy's Office of Scientific and Technical Information.

Reports not in digital format may be purchased by the public from the National Technical Information Service (NTIS):

U.S. Department of Commerce
National Technical Information Service
5301 Shawnee Rd
Alexandria, VA 22312
www.ntis.gov
Phone: (800) 553-NTIS (6847) or (703) 605-6000
Fax: (703) 605-6900
Email: **orders@ntis.gov**

Reports not in digital format are available to DOE and DOE contractors from the Office of Scientific and Technical Information (OSTI):

U.S. Department of Energy
Office of Scientific and Technical Information
P.O. Box 62
Oak Ridge, TN 37831-0062
www.osti.gov
Phone: (865) 576-8401
Fax: (865) 576-5728
Email: **reports@osti.gov**

Disclaimer

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor UChicago Argonne, LLC, nor any of their employees or officers, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of document authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof, Argonne National Laboratory, or UChicago Argonne, LLC.

Development of a mechanistic source term approach for liquid-fueled Molten Salt Reactors

Prepared by
James Jerden¹, David Grabaskas², Matthew Bucknor²

¹Chemical & Fuel Cycle Technologies Division

²Nuclear Engineering Division

Argonne National Laboratory

July 1, 2019

Abstract

In addition to the significant progress being made in accident analyses for some molten salt reactor (MSR) concepts, there remains a need for a thorough, step-by-step source term analyses that includes parametric sensitivity analyses and trial calculations. This report provides a basis for developing an MSR source term model by examining the successful methodology used in the sodium fast reactor (SFR) mechanistic source term work presented by Grabaskas et al., [2016a]. Useful risk analyses tools currently in use for developing MSR source term analyses are also summarized, such as those developed for the European Safety Assessment of the Molten Salt Fast Reactor (SAMOFAR) project. Based on the analyses summarized in this report, a pathway is recommended for a rigorous and technology-neutral MSR source term analysis in the U.S.

This report is submitted in fulfillment of Advanced Reactor Technologies, Molten Salt Reactor Campaign, Salt Chemistry work package AT-19AN04060104, milestone number M4AT-19AN040601043.

Contents

ABSTRACT	I
1 INTRODUCTION	1
2 MECHANISTIC SOURCE TERM FOR ADVANCED REACTORS	4
2.1 MECHANISTIC SOURCE TERM TRIAL CALCULATION APPROACH	4
2.2 RADIONUCLIDES OF INTEREST.....	6
2.3 MECHANISTIC SOURCE TERM TRIAL CALCULATION IMPLEMENTATION.....	8
2.4 SIMPLIFIED MASS BALANCE SOURCE TERM MODEL	18
2.5 SUMMARY OF FINDINGS FROM THE SFR MECHANISTIC SOURCE TERM ANALYSIS.....	19
3 PRELIMINARY SOURCE TERM ANALYSES FOR MOLTEN SALT REACTORS	21
3.1 SUMMARY OF FINDINGS FROM THE SFR MECHANISTIC SOURCE TERM ANALYSIS.....	21
3.1.1 POWER INCREASE (REACTIVITY INITIATED) TRANSIENTS.....	22
3.1.2 FLOW DECREASE ACCIDENTS	22
3.2 SUMMARY OF THE SAFETY ASSESSMENT OF THE MOLTEN SALT FAST REACTOR PROJECT	23
3.2.1 METHODOLOGY OF TRANSIENT SCENARIO IDENTIFICATION.....	24
4.0 RECOMMENDATIONS AND FUTURE WORK.....	27
REFERENCES	29

Acronyms

ANS	American Nuclear Society
ASME	American Society of Mechanical Engineers
BWR	Boiling Water Reactor
CFR	Code of Federal Regulations
DOE	Department of Energy
EBR-II	Experimental Breeder Reactor II
FFTF	Fast Flux Test Facility
IFR	Integral Fast Reactor
INL	Idaho National Laboratory
LBE	Licensing Basis Event
LWR	Light Water Reactor
MST	Mechanistic Source Term
MSR	Molten Salt Reactor
MSBR	Molten Salt Breeder Reactor
MSFR	Molten Salt Fast Reactor
NRC	Nuclear Regulatory Commission
PRA	Probabilistic Risk Assessment
PRISM	Power Reactor Innovative Small Module
PWR	Pressurized Water Reactor
RTDP	Regulatory Technology Development Plan
SAMOFAR	Safety Assessment of the Molten Salt Fast Reactor
SNL	Sandia National Laboratory
SFR	Sodium Fast Reactor
TREAT	Transient Reactor Test Facility

1 Introduction

For over 40 years the U.S. Nuclear Regulatory Commission (NRC) has used postulated “accidental releases of radioactive materials” (source term) as an essential basis for reactor licensing [NRC, 1995]. Source term is specifically defined in 10 CFR § 50.2 as:

“The magnitude and mix of the radionuclides released from the fuel, expressed as fractions of the fission product inventory in the fuel, as well as their physical and chemical form, and the timing of their release.”

The NRC library glossary (<https://www.nrc.gov/reading-rm/basic-ref/glossary.html>) defines source term as:

“Types and amounts of radioactive or hazardous material released to the environment following an accident.”

Source term analyses are currently used for evaluating the consequences of licensing basis events involving normal reactor operations, anticipated off-normal conditions, design basis transients as well as beyond design basis transient scenarios. In addition to their use in reactor licensing, source term analyses can play a key role in reactor siting, as well as the development of emergency planning zones and reactor site boundaries [NRC 1993, NRC 2003, NRC 2005]. For new generation advanced reactor concepts such as sodium cooled fast reactors (SFRs) and molten salt reactors (MSRs), source term analyses can also provide valuable feedback into the design process and facilitate risk-informed engineering decisions (e.g., Yoshioka et al., 2012, Grabaskas et al., 2016a, Gérardin et al., 2019).

Traditionally, source term analyses for light water reactors (LWRs) have used conservative bounding assumptions (e.g., NRC, 1995). However, for a source term analysis to provide useful feedback into the reactor design process and for it to be convincing as a justification for minimizing the size of reactor site emergency planning zones, it must involve more realistic (i.e., mechanistic) models that do not require conservative assumptions regarding radionuclide release during postulated accidents/transients.

The NRC has recognized the usefulness and need for more realistic source term analyses (mechanistic source term or MST) since the 1990s (e.g., NRC, 1993; NRC, 2003; NRC, 2005). While no formal definition for mechanistic source term has been established, the NRC has described a MST in SECY-93-092 (NRC, 1993) as:

“...the result of an analysis of fission product release based on the amount of cladding damage, fuel damage, and core damage resulting from the specific accident sequences being evaluated. It is developed using best-estimate phenomenological models of the transport of the fission products from the fuel through the reactor coolant system, through all holdup volumes and barriers, taking into account mitigation features, and finally, into the environs.”

Such models are referred to as mechanistic because they take into account the real transport/retention processes (e.g., solubility, precipitation, vaporization, adsorption, aerosolization) based on fundamental chemistry (e.g., thermodynamics, electrochemistry, kinetics).

The NRC staff recommendations for new license applications for advanced reactors that were documented in SECY-93-092 further established the essential characteristics of a mechanistic source term (NRC, 1993):

“...source terms should be based upon a mechanistic analysis and will be based on the staff’s assurance that the provisions of the following three items are met:

- *The performance of the reactor and fuel under normal and off-normal conditions is sufficiently well understood to permit a mechanistic analysis. Sufficient data should exist on the reactor and fuel performance through the research, development, and testing programs to provide adequate confidence in the mechanistic approach.*
- *The transport of fission products can be adequately modeled for all barriers and pathways to the environs, including specific consideration of containment design. The calculations should be as realistic as possible so that the values and limitations of any mechanisms or barrier are not obscured.*
- *The events considered in the analyses to develop the set of source terms for each design are selected to bound severe accidents and design-dependent uncertainties.”*

The mechanistic source term concept was further refined in 2013 with the release of the joint American Society of Mechanical Engineers (ASME) and the American Nuclear Society (ANS) consensus standard “Probabilistic Risk Assessment Standard for Advanced Non-LWR Nuclear Power Plants” (ASME/ANS, 2013). This standard proposes a methodology and technical requirements for performing probabilistic risk assessments (PRAs) used to develop risk-informed decisions for advanced reactor analyses. The standard is necessarily technology neutral so that it covers a broad scope of PRA applications, including the quantification of mechanistic source term analyses (Grabaskas et al., 2015). The standard identifies the following qualitative objectives for mechanistic source term (ASME/ANS, 2013):

- Identification of inventories available for release within the reactor coolant system pressure boundary,
- Identification and characterization of the phenomena affecting radionuclide transport,
- Definition of reactor-specific release categories for use in end state and event sequence grouping,
- Determination of release parameters (e.g. chemical phase, release timing and duration, etc.),
- Identification and evaluation of relevant uncertainties, and
- Documentation of the mechanistic analysis.

The set of high-level requirements for a mechanistic source term identified in the standard (ASME/ANS, 2013) are as follows:

- Release categories shall be defined for defining event sequence end states and for grouping event sequences and event sequence families with the same or similar mechanistic source terms.
- The mechanistic source term analysis shall include a method for determining the mechanistic source term for each release category.
- The mechanistic source term analysis shall include calculations to quantitatively characterize the mechanistic source terms for each release category.
- Uncertainties in the mechanistic source terms and associated radionuclide transport phenomena shall be characterized and quantified to the extent practical. Key sources of model uncertainty and assumptions shall be identified, and their potential impact on the results shall be understood. Those sources of uncertainty that are not quantified shall be addressed via sensitivity analysis.
- The mechanistic source term analysis shall be documented consistent with the applicable supporting requirements.

For a thorough review of the history of source term from a regulatory perspective in the U.S., see Section 2.2 of Grabaskas et al., 2015.

The primary challenge for the mechanistic source term approach for non-LWR advanced reactor concepts is that it likely requires the development of new modeling tools (or the significant alteration of existing codes) and may require a significant experimental program to obtain the fundamental chemical and transport data needed to implement the mechanistic models. Grabaskas et al., [2016a] successfully demonstrated a mechanistic source term approach for a generic metal fueled SFR concept. This SFR mechanistic source term study identified the key data gaps and uncertainties stemming from the lack of knowledge regarding fundamental radionuclide transport processes and provides an excellent example on

which to base future non-LWR advanced reactor source term studies. Such an analyses has not yet been performed for MSR concepts. However, there have been preliminary source term studies that identify the key conceptual issues that will need to be addressed in a scenario-specific MSR source term analyses (e.g., Yoshioka et al., 2012; Gérardin et al., 2019). The following summarizes the key conceptual findings of these early advanced reactor source term studies and, based on these studies, provides recommendations for developing a rigorous source term analyses strategy for MSRs.

2 Mechanistic Source Term for Advanced Reactors

The SFR source term project led by Argonne National Laboratory (Argonne) represents one the most thorough source term studies for advanced reactors that is available in the open literature [Grabaskas et al., 2015; Grabaskas et al., 2016a; Grabaskas et al., 2016b]. In particular, the step-by-step analyses detailed in Grabaskas et al., [2016a] provides an excellent basis on which to plan the development of a source term methodology for MSRs. For this reason, the following section will summarize in some detail the steps followed in that study.

The SFR source term studies led by Argonne, which were performed under the auspices of the U.S. Department of Energy (DOE) Regulatory Technology Development Plan (RTDP), involved three major stages. The first stage involved an information gap analysis investigating impediments to licensing SFRs in the United States (Grabaskas et al., 2015). That analysis concluded that the development of mechanistic source term was the only major topic to have both high regulatory importance and possible “long” lead-time to resolution (Grabaskas, 2015). Based on this conclusion, the second stage involved developing a conceptual plan for advanced reactor source term analyses. This plan involved identifying and characterizing radionuclide sources, identifying and characterizing potential radionuclide transport pathways and phenomena, modeling radionuclide transport pathways and phenomena, and applying these models to evaluate particular reactor transient scenarios (Figure 1). The third stage involved performing the activities summarized in the first four boxes of Figure 1 by assembling the needed computational tools and performing a set of trial calculations. The primary purpose of the trial runs was to determine the adequacy of current computational tools and to elucidate gaps in the knowledge base needed to perform a rigorous mechanistic source term analysis for SFRs (Grabaskas, 2016).

Based on the success of these SFR studies, it seems likely that using a similar three stage conceptual approach could be used to expedite developing a source term strategy for MSRs. Therefore, implementation of the SFR approach will be discussed.

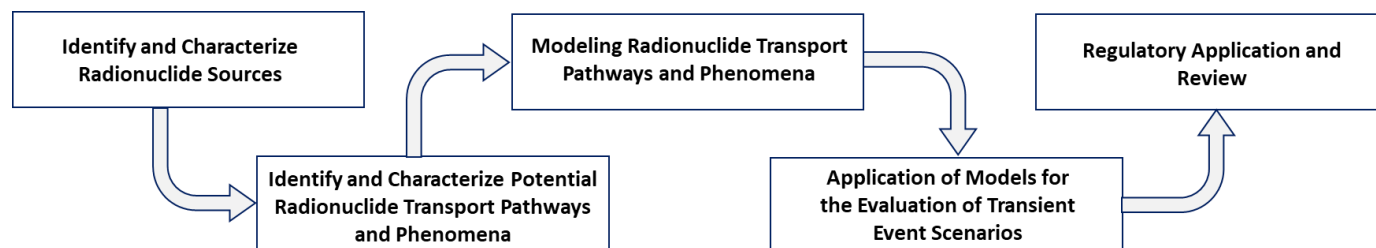


Figure 1. Advanced reactors source term development pathway for SFRs (adapted from Figure 1-1 Grabaskas et al., 2016a). The light blue boxes indicate activities discussed in Grabaskas et al., 2015, the green box is the focus of Grabaskas et al, 2016 and the gray boxes indicate on-going or future work.

2.1 Mechanistic Source Term Trial Calculation Approach

The SFR trial calculations were performed in two parallel paths (Figure 2). For the primary path, which was performed by staff at Argonne and Sandia National Laboratories, best-estimate models and codes were used to perform as realistic-as-possible predictive assessment of the transport and release of radionuclides from a damaged SFR core. Again, the overall goal of this assessment was to identify gaps in computational capabilities or the associated knowledge base [Grabaskas, 2016a]. The other path, performed by independent contractor Dr. R. Denning, was a simplified sensitivity calculation based on a mass balance approach that treated each stage of the source term progression as consisting of a set of independent first-order processes and utilized sets of bounding assumptions (e.g., leak rates from barriers and hold-up volumes). This simplified model provides a parametric evaluation of key radionuclide transport and

retention phenomena in terms of their importance to off-site dose and land contamination consequences [Grabaskas, 2016a].

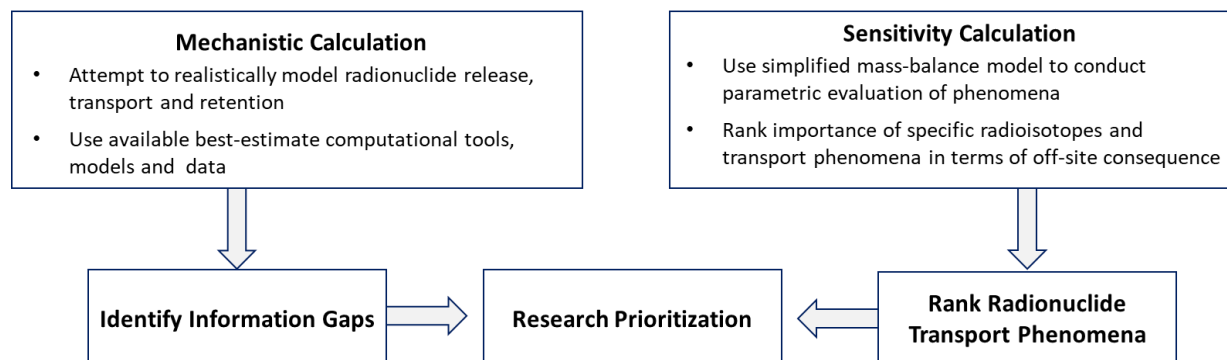


Figure 2. Project methodology for SFR source term study (adapted from Figure 1-3 Grabaskas et al. 2016a).

In order for the mechanistic trial calculations to provide an adequate assessment of the suitability of existing tools and data for future licensing applications, the study had to include three essential characteristics [adapted from Grabaskas, 2016a]:

1. The approach attempted to account for all pertinent radionuclide transport and retention phenomena. When it was discovered that existing models could not account for a particular phenomenon, this was noted as a capability or information gap and conservative assumptions were made with regards to its influence on the calculation. The compilation of gaps identified by this process was used to recommend future work in code development and experimental measurements.
2. The modeling tools used needed to be similar to those typically used in nuclear safety analyses. That is, the codes should not require a super computer to run and should not involve unreasonably long execution times.
3. The design of the reactor used to define the transient scenario examples needed to be representative of designs currently being considered in the SFR industry, particularly with respect to radionuclide transport barriers, hold-up volumes and potential release pathways.

The order of the tasks performed for the mechanistic source term trial calculations for SFRs are shown in Figure 3. Each of these steps are generic enough to also apply to other advanced reactors such as MSRs. The following discussion will highlight the key generic considerations that must be made in each of these steps to successfully implement a mechanistic source term analysis.

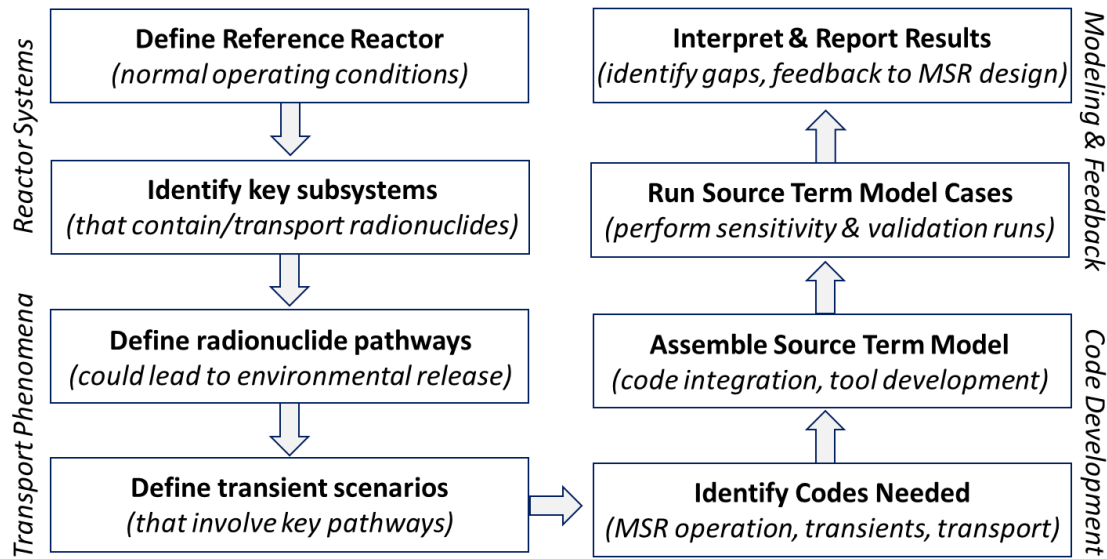


Figure 3. Generic steps involved in the implementation of a mechanistic source term analysis.

2.2 Radionuclides of Interest

Similar to other advanced reactors, the three main sources of radionuclides within the SFR system are:

- Coolant activation products,
- Activated corrosion products from reactor component alloys (Table 1),
- Fission products and actinides present within the fuel (from ORIGEN calculations).

The main activation product within the SFR coolant is Na-24 with a 15-hour half-life; smaller amounts of Na-22 with a 2.6-year half-life are also produced. The short half-life of Na-24 and the low levels of Na-22 produced make sodium activation non-problematic for normal reactor operations (Grabaskas et al., 2015). Furthermore, impurities in reactor-grade sodium are not considered to have significant impact on activity from activation products within the coolant (IAEA, 1993). A similar screening for activation products and the role of possible impurities needs to be performed for MSRs.

Table 1. Activation products that may be released to coolant during steel corrosion.

Nuclide	Progenitors	Half-life (days)	Comments
⁵¹ Cr	⁵⁰ Cr, ⁵² Cr	27.7	Low yield activation product in steels, low impact on source term
⁵⁴ Mn	⁵⁴ Fe, ⁵⁵ Mn	312.3	Most significant corrosion nuclide, could be the major source of activity in sodium, in the absence of radionuclides released from fuel.
⁵⁸ Co	⁵⁸ Ni, ⁵⁸ Co	70.86	Associated with Co impurities and Ni alloys
⁵⁹ Fe	⁵⁸ Fe, ⁵⁹ Co	44.5	Low yield activation product in steels, low impact on source term
⁶⁰ Co	⁶⁰ Ni, ⁵⁹ Co	1925.1	Second most important corrosion nuclide, source is Co impurities in steels and Ni alloys
⁶⁵ Zn	⁶⁴ Zn	244.3	Can be important dose contributor due to activity and chemistry – can be eliminated by eliminating all Zn-bearing materials (e.g., rust proofing)
^{110m} Ag	¹⁰⁹ Ar	249.8	Suspected source is Ag impurity in Ni.
¹⁸² Ta	¹⁸¹ Ta	114.4	Source concentration increases if Nb-bearing steels and alloys are used

Tritium will be present in the sodium coolant as it is produced by ternary fission in the core but readily migrates through fuel cladding and steel structures at operating temperatures (Osterhout, 1978). Tritium is generally not considered an operational concern; its distribution throughout the plant is significantly dependent on the reactor operating conditions and layout (Grabaskas et al., 2015). In an SFR, the majority of oxygen and hydrogen within the sodium coolant are captured during a single pass through a cold trap

(Grabaskas et al., 2015). Operational experience from EBR-II showed that around 45 – 85% of the tritium present in the sodium was also removed per pass through the cold trap (Osterhout, 1978). Tritium is generally not considered a major radionuclide of concern due to its low activity relative to other prominent gaseous radionuclides such as Ne-23 and Ar-41.

In general, LWR source term studies group radionuclides into elemental categories (Table 2) based on similar chemical behavior. This approach is convenient for traditional bounding source term studies; however, as discussed in Section 3 of Grabaskas et al., [2016a], more detailed source term studies quantify the importance and behavior of individual radioisotopes. The consideration of individual isotopes allows the source term study to show which radionuclide transport and retention processes have the most significant impact on the off-site dose consequence for different reactor transient scenarios. Furthermore, grouping radionuclides by their chemical behavior in LWR type systems may lead to misleading results for the non-aqueous environments present in SFRs and MSR.

Table 2. Radionuclide groups specified in Table 3.8 of NUREG-1465.

Element Group #	Element Group	Elements in Group
(NUREG-1465)		
1	Noble Gases	Xe, Kr
2	Halogens	I, Br
3	Alkali Metals	Cs, Rb
4	Tellurium Group	Te, Sb, Se
5	Barium, Strontium	Ba, Sr
6	Noble Metals	Ru, Rh, Pd, Mo, Tc, Co
7	Lanthanides	La, Zr, Nd, Eu, Nb, Pm, Pr, Sm, Y, Cm, Am
8	Cerium Group	Ce, Pu, Np

Therefore, the SFR mechanistic source term analyses tracked around 750 radionuclides for each transient scenario. The tracked inventories, determined by ORIGEN calculations, included some stable isotopes that were suspected to play important roles in chemical speciation and bubble formation behavior [Grabaskas et al., 2016a].

The tracked radionuclide inventories were weighted based on inhalation dose for dose consequence sensitivity analyses. The weighting scheme involved the use of the NRC RASCAL code and assumed 10^8 Ci of each radioisotope was released over a short time period and the dose calculated three miles from the release point assuming a wind speed of 5 meters per second [Grabaskas et al., 2016a]. The assumed distance from the radionuclide release point and meteorological assumptions would significantly affect the absolute doses calculated in this weighing model, but the relative doses between isotopes is not influenced by these assumptions [Grabaskas et al., 2016a]. Therefore, the weighing model produces a reliable ranking of the importance of individual radioisotopes for off-site dose consequences as quantified by the total effective dose equivalent (TEDE). An example dose-weighted inventory from Grabaskas et al., [2016a] is shown in Figure 4. In the example shown in Figure 4, the high dose consequence ranking of plutonium, americium and curium isotopes is due to the very large inhalation doses associated with these alpha-emitters.

Another key point to be considered when selecting the radionuclides to track for a given source term analyses is the possible importance of land contamination. A significant insight from the Fukushima accident was the importance of land contamination and associated relocation of members of the public and decontamination costs [Grabaskas et al., 2016a]. Longer-lived isotopes that may be insignificant for short-term dose consequence calculations could have high importance with respect to land contamination and should be tracked within the source term analyses [Grabaskas et al., 2016a].

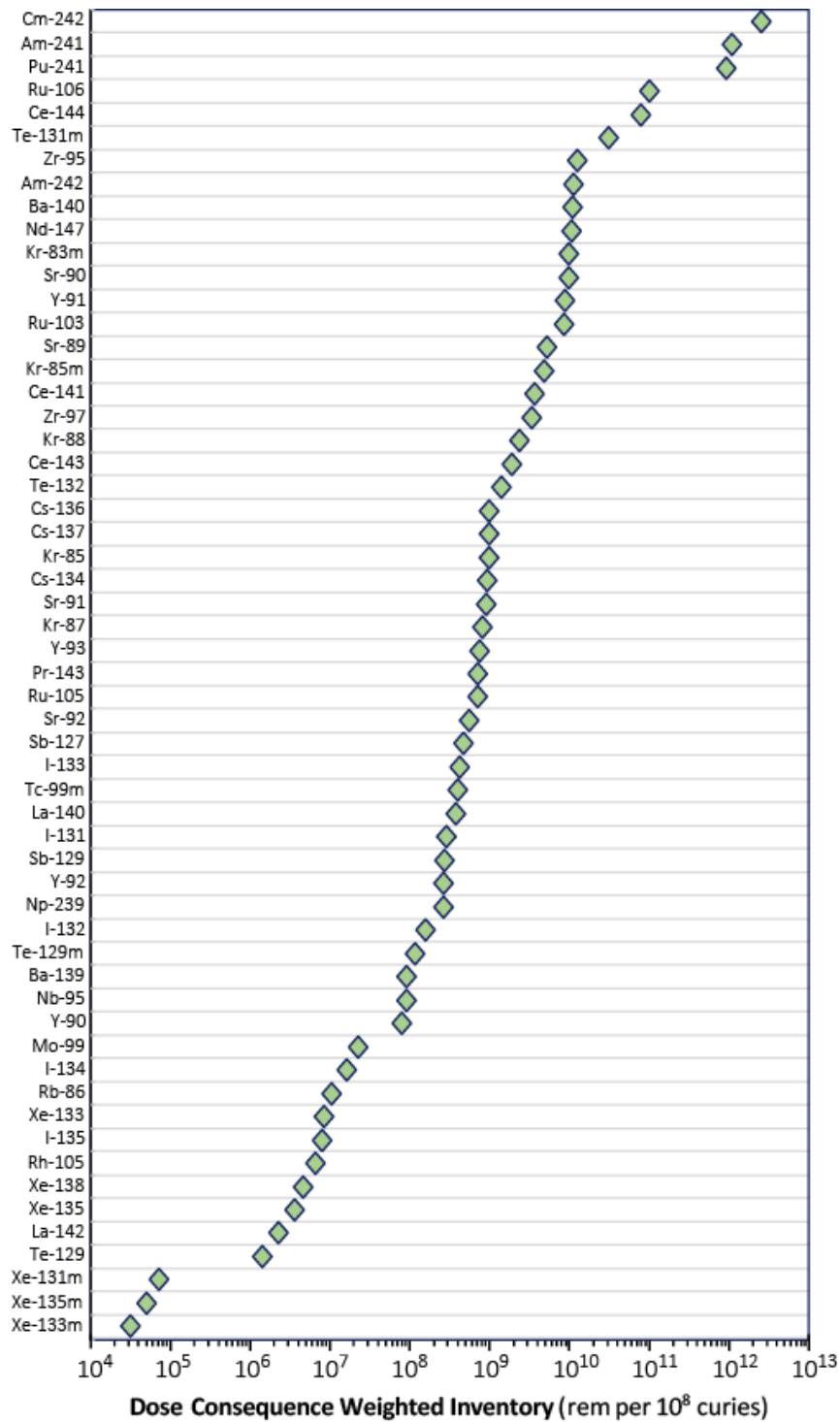


Figure 4. Example dose consequence weighted inventory in rem per 10⁸ curies. This inventory is for an actinide burner SFR [see Section 3.0 in Grabaskas et al., 2016a].

2.3 Mechanistic Source Term Trial Calculation Implementation

As stated above, the key to the first step in the source term analysis implementation is to define a reference reactor that is representative of industry reactor concepts with respect to radionuclide barriers and transport pathways. Table 3 shows the main specifications of the SFR reference design used for the mechanistic source term study, which provides an example of the level of detail that may be required for this step.

Table 3. Characteristics of reference SFR used for the mechanistic source term analyses (adapted from Grabaskas et al., 2016a).

Parameter	Description
Reactor Power	1000 MWth/380MWe
Type	Pool-type
Purpose	Actinide Burner
Core Fuel	U-Pu-Zr
Core Fuel Batches	3
Fuel Burnup Range (at%)	2% – 10%
Fuel Pin Pressure Range (MPa)	0.5 (low burnup) – 6.7 (high burnup)
Primary Coolant Temp. (inlet/outlet)	350°C/500°C
Number of Fuel Assemblies	180
Fuel Pins per Assembly	271

The identification of key reactor subsystems (e.g., radionuclide transport barriers and hold-up volumes) is done in parallel with the identification of radionuclide pathways. For example, Figure 5. shows the main generic radionuclide release pathway through the major reactor subsystems that act as barriers to release. For the SFR example in Figure 5(a), the generic pathway involves the following [Grabaskas et al., 2016a]:

- Release from damaged fuel the fuel into the coolant pool.
- Release from the coolant pool to the cover gas plenum by vaporization or aerosol transport.
 - The coolant pool barrier may be bypassed by bubble migration involving the transport of aerosols and vapor species within krypton/xenon bubbles formed during fuel pin failure.
- Release from the cover gas plenum into the containment volume through the leakage of aerosols and vapor species.
- Release from the containment volume to the environment through the leakage of aerosols and vapor species.

A hypothetical release pathway for a generic MSR is shown in Figure 5(b) for comparison; this example is considered preliminary as a detailed MSR source term analyses has not yet been performed. Figure 6 displays more of the details of processes that determine the distribution and transport of radionuclides during reactor operation. The processes shown in Figure 6 need to be modeled in the source term analyses to predict the masses of radionuclides released through the generic pathways identified in Figure 5. In Figure 6(a), the example given for the MSR case is again preliminary and is presented here to provide a sense of some of the generic similarities and differences between the SFRs and MSRs with regard to source term.

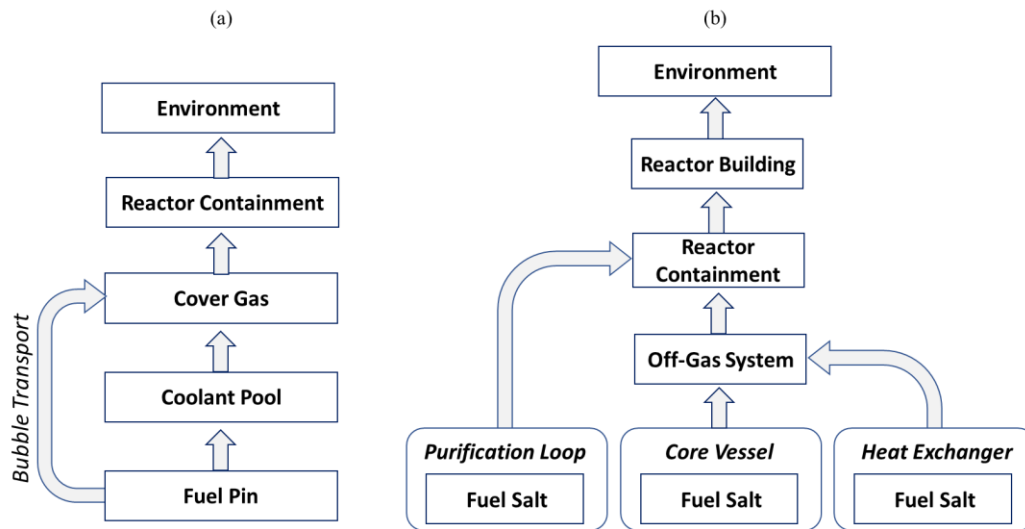


Figure 5. Conceptual diagram identifying the main release pathways for radionuclides in a generic SFR [e.g., Grabaskas et al., 2016a] (a) and a generic MSR (b).

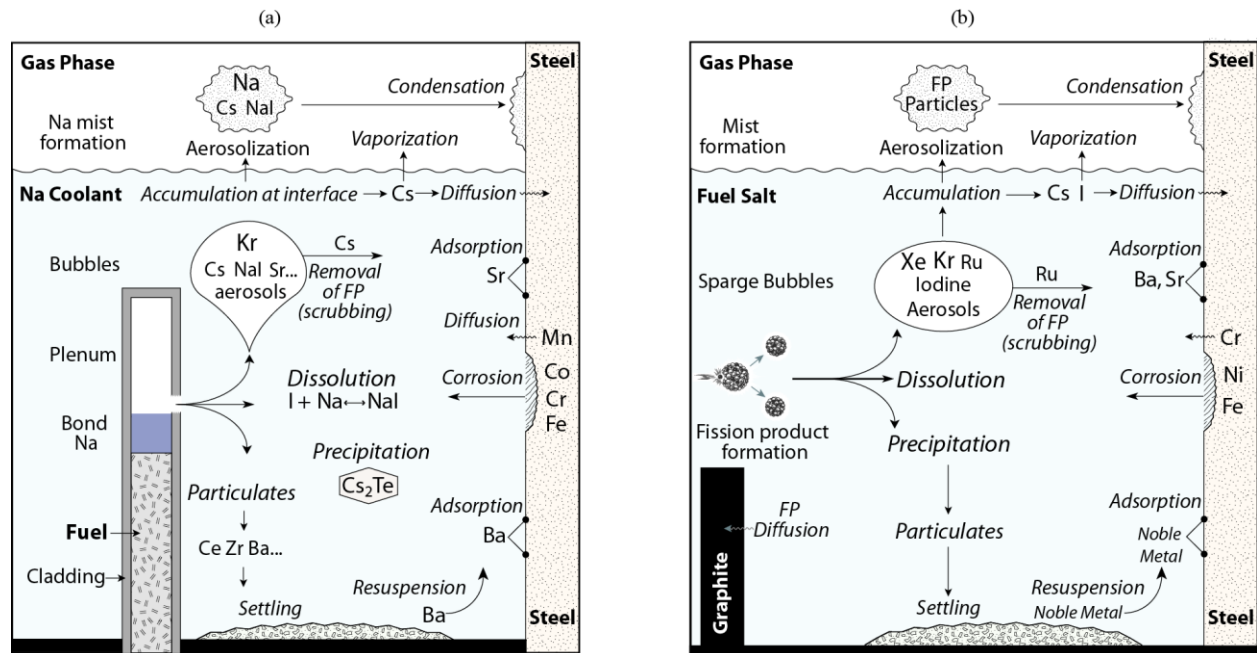


Figure 6. Schematic diagram identifying processes that determine the distribution of radionuclides in a generic SFR (a) and a generic MSR (b). The schematic behavior of elements shown in these diagrams are from limited observations and generic chemical behavior and are largely hypothetical.

After defining the key reactor subsystems and associated radionuclide pathways (Figure 5), transient scenarios that involves those systems and pathways need to be defined. Types of transients can be classified as (Figure 7):

- Abnormal operating transients that include anticipated events such as equipment malfunction and low consequence operator error.
- Design basis accidents which are unanticipated events such as cladding failure in solid fueled reactors.
- Severe accident events involving significant damage (fuel melting) and radionuclide release from the core.

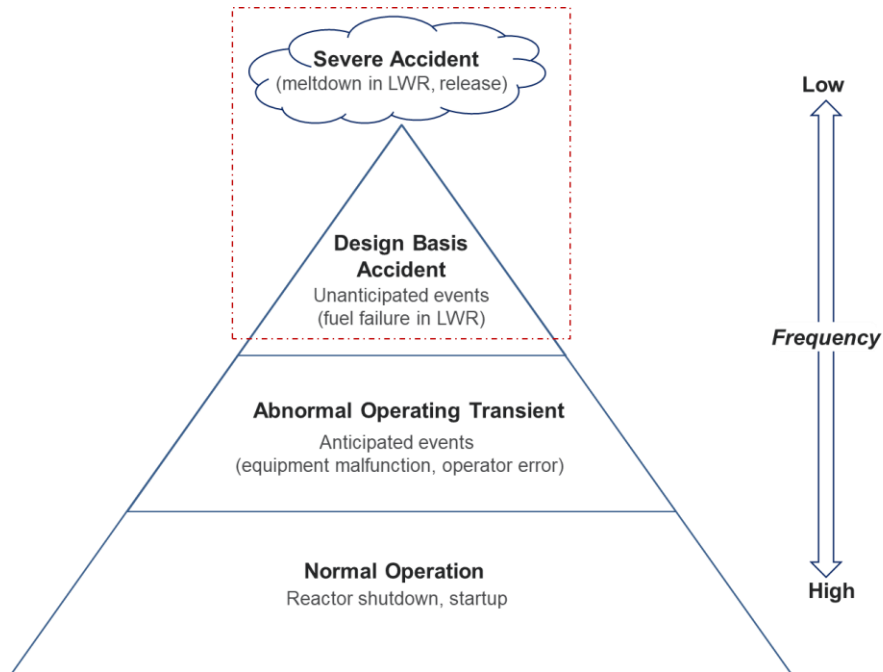


Figure 7. Summary categories of reactor transient and accident scenarios (adapted from Yoshioka et al., 2012). The red dotted line indicates the categorization of the transients investigated for the SFR source term study [Grabaskas et al., 2016a].

For the SFR source term study, two types of accident scenarios were defined based on the need to provide the type of core damage conditions that would allow for a thorough assessment of all relevant radionuclide pathways as well as provide an adequate assessment of the computational tools. As emphasized in Grabaskas et al., [2016a] it is important at this stage of the source term analyses to be clear about the reasons particular transient or accident scenarios were chosen. As mentioned, the accident scenarios for the SFR case were chosen to facilitate a thorough assessment of capabilities and it was emphasized that the selection of the specific transients should not be misconstrued as an indication of their particular importance to future SFR licensing efforts [Grabaskas et al., 2016a].

To provide an example of the level of detail needed for this stage of the source term analyses, the assumed transient scenario characteristics and timelines used for the SFR study are shown in Table 4.

Table 4. Transient scenarios assumed for the SFR source term study [Grabaskas et al., 2016a].

Protected Loss of Flow with Degraded Decay Heat Removal		Severe Unprotected Transient Overpower	
Slow rise of Core Temperatures Fuel pins fail due to eutectic penetration, no bulk fuel melting Very high primary system temperatures		Rapid rise in core temperatures Fuel melting resulting in pin failures Near-nominal primary system conditions	
Time	Event	Time	Event
0 sec	Loss of primary flow (successful coastdown)	0 sec	Reactivity insertion (\$0.075 per sec)
0 sec	Loss of balance of plant heat removal	2 sec	Reactor SCRAM fails
0 sec	Degraded DRACS	11.5 sec	End of SAS4A/SASSYS-1 analysis
2 sec	Reactor SCRAM		
72 hrs	Decay heat removal restored		
168 hrs	End of SAS4A/SASSYS-1 analysis		

The Argonne-designed SFR-specific code SAS4A/SASSYS-1 was used to quantify reactor conditions during the transients described in Table 4. This code was developed for the deterministic analyses of both design basis and beyond design basis events and includes mechanistic models for both steady-state and transient thermal-hydraulic, neutronic and mechanical phenomena. For the mechanistic SFR source term, calculations using the SAS4A/SASSYS-1 code quantified the following characteristics needed for propagating the source term [Grabaskas et al., 2016a]:

- Time of fuel failure.
- Fuel temperature at failure.
- Core coolant pool temperature at failure.
- Internal fuel pin pressure immediately prior to failure.
- Number of fuel pins failed.

The models constituting the SAS4A/SASSYS-1 code were validated with analyses of reactor and plant data from the EBR-II, FFTF and TREAT reactors (e.g., Thomas et al., 2012).

Relational diagrams linking inputs and outputs of codes and calculations were used to organize the flow of information throughout the source term calculation. Figure 8 shows the input-output diagram for the transient scenario modeling. This practice of using relational diagrams that clearly communicate the flow of information at each stage of the source term calculation was found to be successful and is recommended as a practice for the MSR source term development efforts.

**Figure 8.** Relational diagram showing the flow of inputs and outputs for the transient scenario modeling performed for the SFR source term study [adapted from Grabaskas et al., 2016a].

After establishing the conditions in the core during the transient scenarios, the radionuclide distribution within the fuel pins (i.e., partitioning between bond sodium, plenum gas and solid) was determined using literature data, ORIGEN calculations, and the thermochemical modeling code HSC chemistry [Grabaskas et al., 2016a]. These inputs and outputs are summarized in Figure 9.

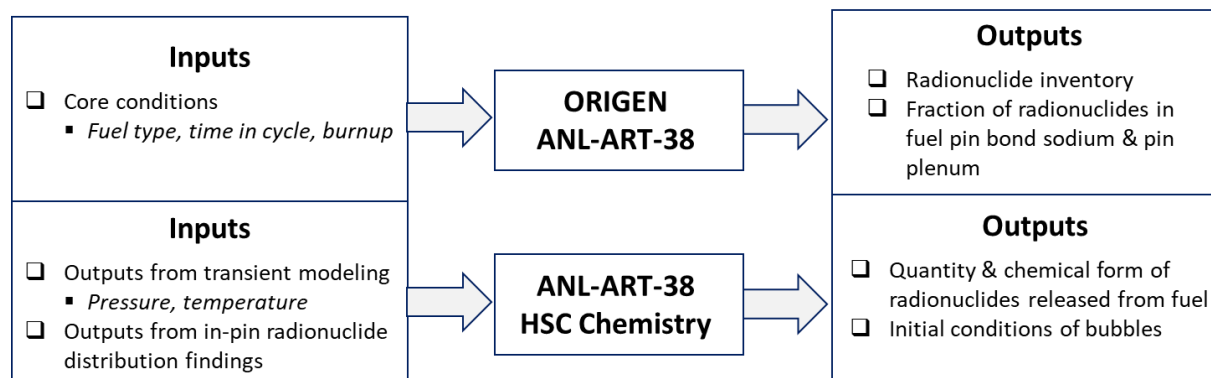


Figure 9. Relational diagram showing the flow of inputs and outputs for the radionuclide distribution calculations performed as part of the SFR source term study [adapted from Grabaskas et al., 2016a]. Note: ANL-ART-38 refers to the report Grabaskas et al., [2016b].

Because the fuel pins are pressurized, their failure leads to a “blow-out” of plenum gas (mainly xenon and krypton), bond sodium and possibly fuel particulates [see Figure 6(a)]. The noble gases form bubbles that are conceivably capable of transporting aerosols and vapor species. It was particularly important in the SFR analyses to determine the types and quantities of radionuclides transported as aerosols or vapor species within the noble gas bubbles because this pathway by-passes the sodium coolant pool, which is one of the major barriers to radionuclide release [Grabaskas et al., 2016a]. One of the major uncertainties for the SFR source term case was the amount of radionuclides present in the noble gas bubbles at pin failure. This quantity was bounded by assuming, in one case, that all radionuclides present in the fuel pin bond sodium and plenum become entrained in the bubbles and, in another case, that only 10% of radionuclides present in the bond and plenum were entrained in the bubbles [Grabaskas et al., 2016a].

As the noble gas bubbles migrate from the core, through the sodium coolant and up to the cover gas region, entrained radionuclides may be removed (scrubbed) from the bubbles by several processes. This is an important radionuclide mass balance issue because entrained fission products and activation products are continuously deposited from the gas bubble into the sodium coolant during bubble migration [Grabaskas et al., 2016a]. The dominant processes involved in gas scrubbing are summarized in Table 5.

Table 5. Processes associated with the removal of aerosols from bubbles during migration through the coolant [adapted from Grabaskas et al., 2016a].

Phenomenon	Description	Modeled in IFR Bubble Code
Brownian Diffusion	Random movement of aerosol within a fluid	Yes
Inertial Deposition	Impaction of aerosol due to inertia	Yes
Gravitational Sedimentation	Settling of aerosol due to gravity	Yes
Vapor Condensation (Na)	Aerosol removal due to Na vapor condensation	Yes
Vapor Condensation (non-Na)	Aerosol removal due to vapor condensation (non-Na)	No
Thermophoresis	Motion of aerosol due to temperature gradient	No
Diffusiophoresis	Motion of aerosol due to diffusion gradient	No

These processes were quantified in the SFR analyses using a code developed at Argonne as part of the IFR project (unpublished work 1995). The code uses classical theories of aerosol and vapor species scrubbing (involving the processes listed in Table 5) to determine the quantities of radionuclides removed from the bubbles and deposited into the sodium coolant during transport from the core to the cover gas. The inputs and outputs for the IFR bubble code are shown in Figure 10.

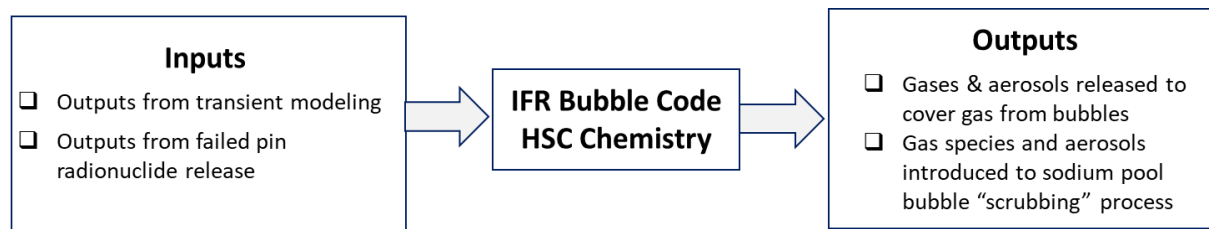


Figure 10. Relational diagram showing the flow of inputs and outputs for the bubble transport calculations performed as part of the SFR source term study [adapted from Grabaskas et al., 2016a].

Although bubble generation in a liquid fueled MSR will obviously be quite different from the SFR scenario, radionuclide entrainment in bubbles is anticipated to play an important role in the distribution of fission products during MSR operations and possible transient scenarios. Some MSR designs (e.g., Yoshioka et al., 2012; Gérardin et al., 2019) include a gas sparge process in which helium or another inert gas is injected into the fuel salt to “sweep” or capture noble gas fission products and possibly some condensed phases such as noble metals and remove them to an off-gas treatment system. A code similar to the IFR bubble code will be needed to quantify the amount of non-noble gas radionuclides that will be transported by the sweep-gas bubbles for the MSR source term analyses.

Besides the bubble transport pathway, the main process leading to radionuclide transport from the sodium coolant to the cover gas region is volatilization at the coolant/gas interface. Modeling this process requires the calculation of radionuclide vapor pressures over the radionuclide-bearing coolant solution (molten sodium for the SFR case). This calculation requires knowledge of the chemical speciations of the radionuclides of interest and the activities of those species within the coolant solution. For the SFR source term analyses, the vaporization process was modeled using the thermodynamic code HSC Chemistry. For this analysis, a custom thermodynamic database (including activity coefficients) was compiled from the literature and used by the HSC code to determine the equilibrium speciation of key species (e.g., Cs, I, NaI, CsI) for each step in the transient scenarios [Grabaskas et al., 2016a]. The compilation of the custom HSC database is discussed in detail in Appendix C of Grabaskas et al., 2016a and summarized in Jerden et al., 2019. The flow of information involved in the implementation of this chemistry model for the SFR source term study is shown in Figure 11 and the inputs and outputs are summarized in Figure 12.

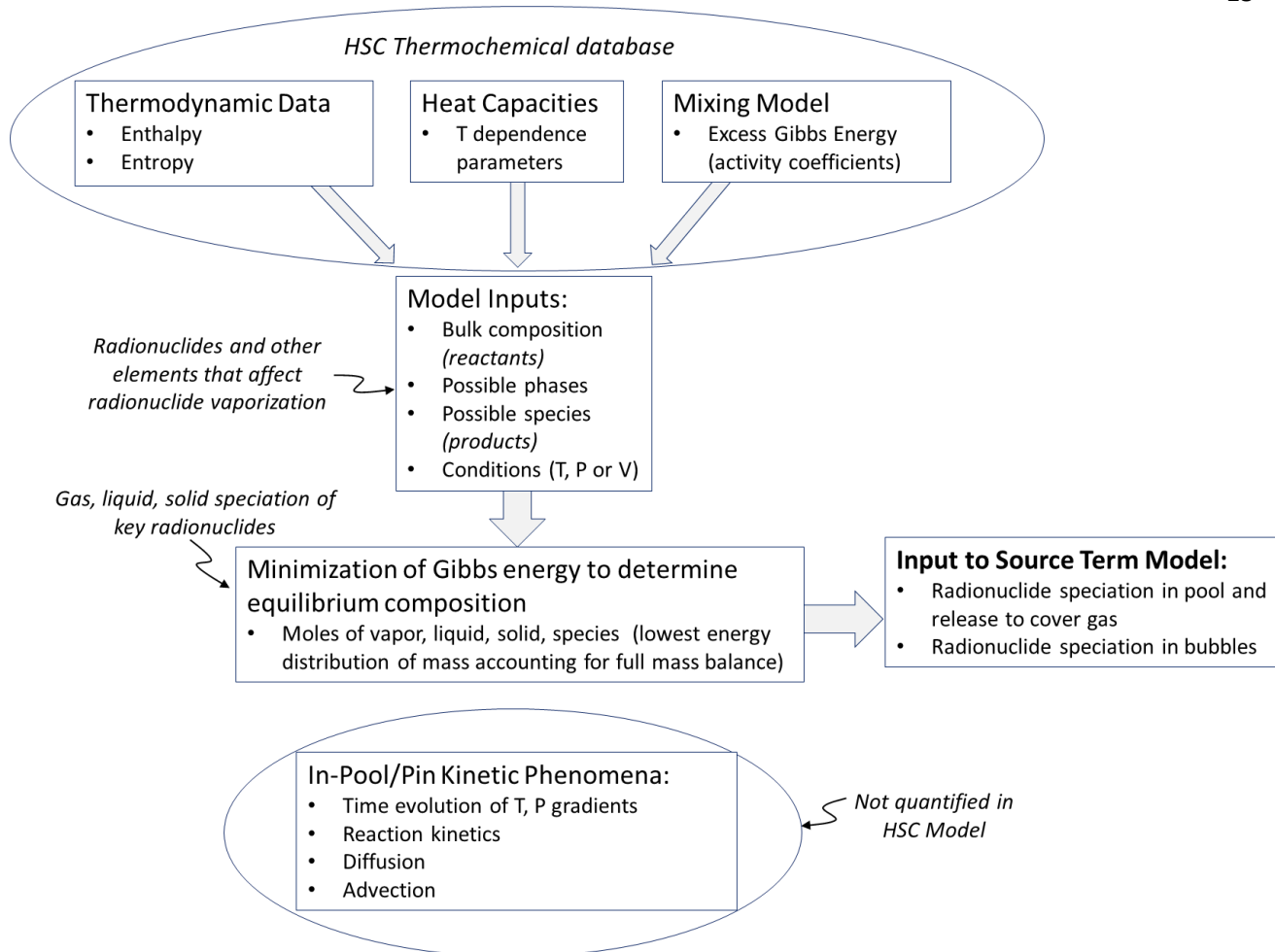


Figure 11. Information flow involved in the development and implementation of the chemical speciation model used for the SFR source term analyses. For details see Appendix C of Grabaskas et al., 2016a.

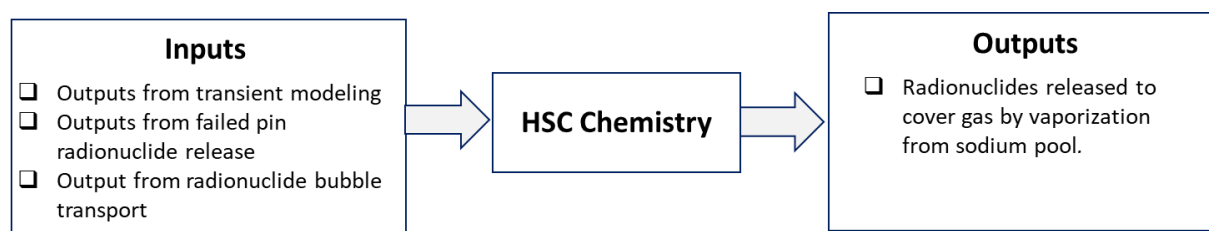


Figure 12. Relational diagram showing the flow of inputs and outputs for the chemical speciation calculations performed as part of the SFR source term study [adapted from Grabaskas et al., 2016a].

A chemical speciation model will also be needed for the MSR source term study to determine the distribution of radionuclides between the vapor and condensed phases. The importance of chemical speciation to radionuclide transport and source term analyses for MSRs is discussed in McFarlane et al., 2018.

In addition to thermodynamic equilibrium vaporization, kinetic process may also play an important role in determining the distribution of radionuclides between the coolant liquid and cover gas. The kinetics of vaporization are influenced by the coolant fluid dynamics, the liquid surface area and other geometry-specific factors. The key factors that are and are not accounted for in the HSC chemistry model are shown in Table 6. Since most of these kinetic factors are poorly quantified and unknown for the particular SFR

transient scenarios under investigation, they were not quantified as part of the HSC chemistry model (see Figure 9). The model is deemed to be conservative in that it assumes instantaneous equilibration and ignores processes that could lead to the retention of radionuclides within the coolant, such as plate-out, gravitational settling, and localized oxidation (see Table 4).

Table 6. Processes associated with the transport of radionuclides from the coolant to the cover gas regions [adapted from Grabaskas et al., 2016a]

Phenomenon	Description	Modeled in HSC Calc.
Thermodynamics	Vapor pressure of specific chemical species	Yes
Non-ideal Mixing	Activity coefficient corrected vapor pressures of chemical species	Yes
Kinetic Effects	Chemical reaction rates	No
Mechanical Forces	Transport to cover gas due to bubble bursting, spraying, misting	No
Plate-out and Adsorption	Removal of radionuclides in pool due to interactions with structures	No
Gravitational Settling	Removal of radionuclides in pool due to settling	No
Compositional Heterogeneity	Localized concentrations of radionuclides in the pool	No
Localized Oxidation	Creation of oxide film on pool surface, slowing vaporization	No

The transport of radionuclides from the cover gas region to the reactor containment volume was performed using the Simplified Radionuclide Transport Code (SRT), which was recently developed at Argonne for the SFR source term analyses [Grabaskas et al., 2016a]. The SRT code is a zero-dimension, time-based analyses tool that considers three control volumes: the cover gas region, the containment volume and the environment. It assumes perfect mixing in each volume and allows for user-specified leakage rates from the cover gas to containment and from containment to the environment [Grabaskas et al., 2016a]. The code accounts for more than 700 isotopes and quantifies radionuclide transport/retention based on the processes summarized in Table 7. The inputs and outputs for the cover gas retention calculations are summarized in Figure 13. Because of the significant uncertainties regarding the design of the SFR reactor head and the possible leakage rates from the cover gas region into the containment volume, two bounding cases were considered: a 0.55 vol % leak rate per day and a 52 vol % leak rate per day.

Table 7. Processes accounted for in the Simplified Radionuclide Transport Code used to quantify radionuclide distribution between the cover gas and reactor containment for the SFR source term study [adapted from Grabaskas et al., 2016a].

Phenomenon	Description
Condensation	Reduced temperatures may result in condensation of radionuclide vapors.
Deposition	Radionuclide aerosols may settle on structures or pool surface due to inertial and gravitational forces. Radionuclide vapors may adsorb on structural surfaces.
Agglomeration	Radionuclide (and sodium) aerosols may agglomerate, changing their properties.
Decay	Radionuclide decay will occur (only daughters of noble gases are considered in calculation).
Chemical Interactions	Radionuclides may chemically interact with each other and sodium vapor.
Seal Leakage	Seals in the reactor head will leak depending on temperature and pressure.

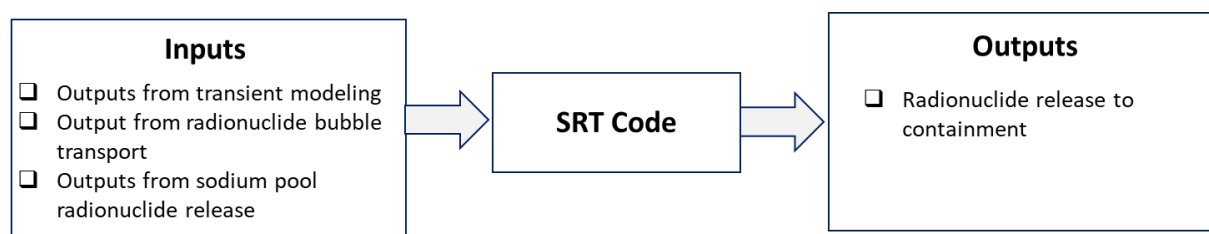


Figure 13. Relational diagram showing the flow of inputs and outputs for the transport of radionuclides from the cover gas region to the reactor containment performed as part of the SFR source term study [adapted from Grabaskas et al., 2016a].

It is anticipated that the same types of processes shown in Table 7 will be applicable for calculating the possible transport of radionuclides from cover gas or plenum volumes to the reactor containment region for MSR transient scenarios. Therefore, a code such as SRT may need to be developed with MSR specific parameters and processes.

The SRT code was also used to quantify the transport of radionuclides from the containment volume into the environment. The radionuclide transport/retention phenomena active within the containment volume are very similar to those accounted for in the cover gas region (Table 7) with the major difference being that radionuclides in the containment volume may interact with water vapor and oxygen from air [Grabaskas et al., 2016a]. This highlights one of the limitations of the SRT code, it is not yet able to account for chemical interactions with oxygen. Perfect mixing was assumed for that analysis, which resulted in the initiation of leakage of radionuclides to the environment immediately following their introduction to containment [Grabaskas et al., 2016a].

The WinMACCS code was used to calculate the dispersion of radionuclides that leak from containment into the environment. This calculation involved quantifying the total effective dose equivalent (TEDE) of a maximally exposed offsite individual at several distances from the reactor site [Grabaskas et al., 2016a]. The selection criteria were based on 10 CFR § 50.34. For regulatory purposes, the TEDE is typically used as the primary human health response measure and provides the basis for determining reactor site exclusion area boundaries [Grabaskas et al., 2016a]. The processes accounted for in this analysis are summarized in Table 8 and the inputs and outputs for this part of the analyses are shown in Figure 14.

Table 8. Processes associated with the transport of radionuclides from reactor containment into the environment [adapted from Grabaskas et al., 2016a]

Phenomenon	Description	Modeled in WinMACCS
Dispersion	Plume dispersion	Yes
Decay	Radionuclide decay	Yes
Daughter Products	Daughter products from decay	Yes
Chemical Interactions	Chemical form of the radionuclide and possible interactions	No
Deposition	Wet/dry deposition	Yes
Meteorology	Weather conditions that impact plume dispersion	Yes
Siting	Site characteristics	Yes
Evacuation	Evacuation of population	Yes
Dose Factors	Dose conversion factors for each radioisotope	Yes



Figure 14. Relational diagram showing the flow of inputs and outputs for the transport of radionuclides from the reactor containment to the environment performed as part of the SFR source term study [adapted from Grabaskas et al., 2016a].

2.4 Simplified Mass Balance Source Term Model

The simplified mass-balance model that was developed and implemented in parallel with the SFR mechanistic source term model provided the capability to perform a large number of sensitivity studies on key variables affecting the source term [Grabaskas et al., 2016a]. Those sensitivity runs allowed for the quantification and ranking of the relative importance of the radionuclide pathways identified during the initial stages of the mechanistic source term study (e.g., Figure 6). Those runs would have been far more time consuming and complicated to perform using the mechanistic source term tool [Grabaskas et al., 2016a].

The simplified model does not perform time-dependent analysis of phenomena taking place during transients, but treats each stage of radionuclide transport/retention as an independent process. Specifically, radionuclide transport between reactor hold-up volumes [identified in Figure 5(a)] was defined as a set of simple first order processes quantified as fractional removal rates [see Appendix B of Grabaskas et al., 2016a]. Differential equations quantifying the mass balance for each hold-up volume were developed and integrated over the duration of the transient events described in Table 4.

The results of the sensitivity calculations quantified which radionuclides and transport/retention phenomena were most significant in terms of total effective off-site dose and land contamination. The ranking of radionuclide importance was done by assigning a dose-weighting factor to each isotope (e.g., Figure 4) in the inventory and then tracking each isotope through the release pathways assumed in the simplified model [Grabaskas et al., 2016a].

The sensitivity runs involved increasing or decreasing the radionuclide retention within certain reactor regions. The following processes were used as sensitivity variables [Grabaskas et al., 2016a]:

- Retention of radionuclides in the fuel,
- Rate of radionuclide removal from bubbles within the sodium coolant,
- Radionuclide aerosol deposition rates,
- Cover gas leakage rate,
- Containment leakage rate.

The findings of these sensitivity run are summarized in Table 9.

Table 9. Summary of rankings of radionuclide transport phenomena from the sensitivity analyses performed for the SFR source term analyses [adapted from Grabaskas et al., 2016a].

Phenomena	Importance
Pool Bypass (bubble transport)	Very High
Fuel Release Fractions (actinides/lanthanides)	High
Aerosol Deposition/Removal	Medium
Reactor Head/Containment Leak Rate	Medium
Pool Vaporization	Low
Noble Gas Decay Chains	Low

2.5 Summary of Findings from the SFR Mechanistic Source Term Analysis

The primary advantage to this mechanistic approach to determining source terms is that it may provide advanced reactor vendors with an opportunity to decrease the size of emergency planning zones and plant sites [Grabaskas et al., 2016a] and can also can provide insights for reactor designers facilitating risk-informed design decisions (Grabaskas et al., 2016a; Gérardin et al., 2019).

The overall finding the SFR study was that it is possible with currently available tools and models to perform mechanistic source term evaluations for SFRs. However, a number of significant gaps in models and data for some key phenomena exist, which could lead to large analyses uncertainties for some relevant transient scenarios [Grabaskas et al., 2016a]. These gaps need to be addressed as the current uncertainties would likely make justifying reduced emergency planning zones or plant site sizes difficult and could impact design decisions that could be dependent on the source term model [Grabaskas et al., 2016a].

This analysis showed that there are significant advantages to performing a two-pronged approach to source term analyses involving two types of modeling tools [Grabaskas et al., 2016a]:

1. A mechanistic source term model that can provide valuable feedback into the design process and possibly justify the reduction of size of emergency planning zones and site boundaries.
2. A simplified mass balance model that can be used to perform sensitivity studies to rank and prioritize the key radionuclide release pathways in terms of the total effective dose equivalent to exposed offsite individuals and possible land contamination.

Results from both the mechanistic model and simplified model were used to identify and rank key information gaps that need to be filled to reduce uncertainties in the SFR source term (Table 10). The initial phase of an MSR source term study should seek to produce a similar table that could be used to guide future research and development.

Table 10. Prioritization of phenomena groups based on the SFR source term analysis [adapted from Grabaskas et al., 2016a].

Ranking	Group	Notes
1	Bubble Transport	Represents a potential mechanism to bypass a major radionuclide barrier (the sodium pool). Very high importance in sensitivity calculations, and direct impact on non-noble gas radionuclide transport. Difficult to determine if analysis assumptions are realistic, and significant impact on subsequent analysis steps.
2	In-pin Migration & Release	Determines initial radionuclide release fractions. High importance in sensitivity calculations. Conservative assumptions are straightforward to apply, but assumptions propagate through subsequent analysis steps, resulting in potentially unrealistic releases of transuranics.
3	Aerosol Behavior	Aerosol deposition/condensation is a significant retention mechanism with medium importance in sensitivity calculations. Data regarding deposition, condensation, and chemical interactions are not as well-established as LWR data. Codes, such as MELCOR, have ability to model phenomena, but data are a necessary input.
4	Holdup/Leakage	The delay in radionuclide releases due to hold-up in the cover gas region and containment. Medium importance in sensitivity calculations. Assumptions are straightforward to apply and characterize, as shown in trial MST calculation where multiple leakage values were assumed
5	Vaporization	Vaporization of radionuclides from the sodium pool. Many gaps in the modelling of phenomena, but was low importance in sensitivity calculations (and in trial MST calculation). However, the relative importance of vaporization could increase, if bubble transport calculations can be shown to be overly conservative.
6	Dispersion	Radionuclide characteristics and dose conversion factors. Although these factors have a direct impact on offsite consequence, the magnitude of their influence is likely lower than the factors related to radionuclide release fractions from the plant.

3 Preliminary Source Term Analyses for Molten Salt Reactors

Accident analyses for MSR were performed in the 1960s as part of the molten salt reactor experiment (MSRE) performed at Oak Ridge National Laboratory (e.g., Haubenreich and Engel, 1962; Beall et al., 1964). However, the applicability of these studies to the development of MSR source term analyses is limited because the focus of the MSRE was not on developing safety or licensing tools but to demonstrate the MSR concept. More recently work by Yoshioka et al., (2012) and Gérardin et al., (2019) have provided studies that take steps towards developing a methodology for MSR source term. In this section, these two studies will be summarized to inform the ongoing developmental work on MSR source term in the U.S.

3.1 Summary of Findings from the SFR Mechanistic Source Term Analysis

In their summary of possible guidelines for MSR accident analyses, Yoshioka et al. (2012) present a number of generic transient scenarios for the molten salt breeder reactor (MSBR) concept (e.g., Shimazu, 1978). The reactor concept and the possible transient scenarios discussed in Yoshioka et al., (2012) are shown schematically in Figure 15.

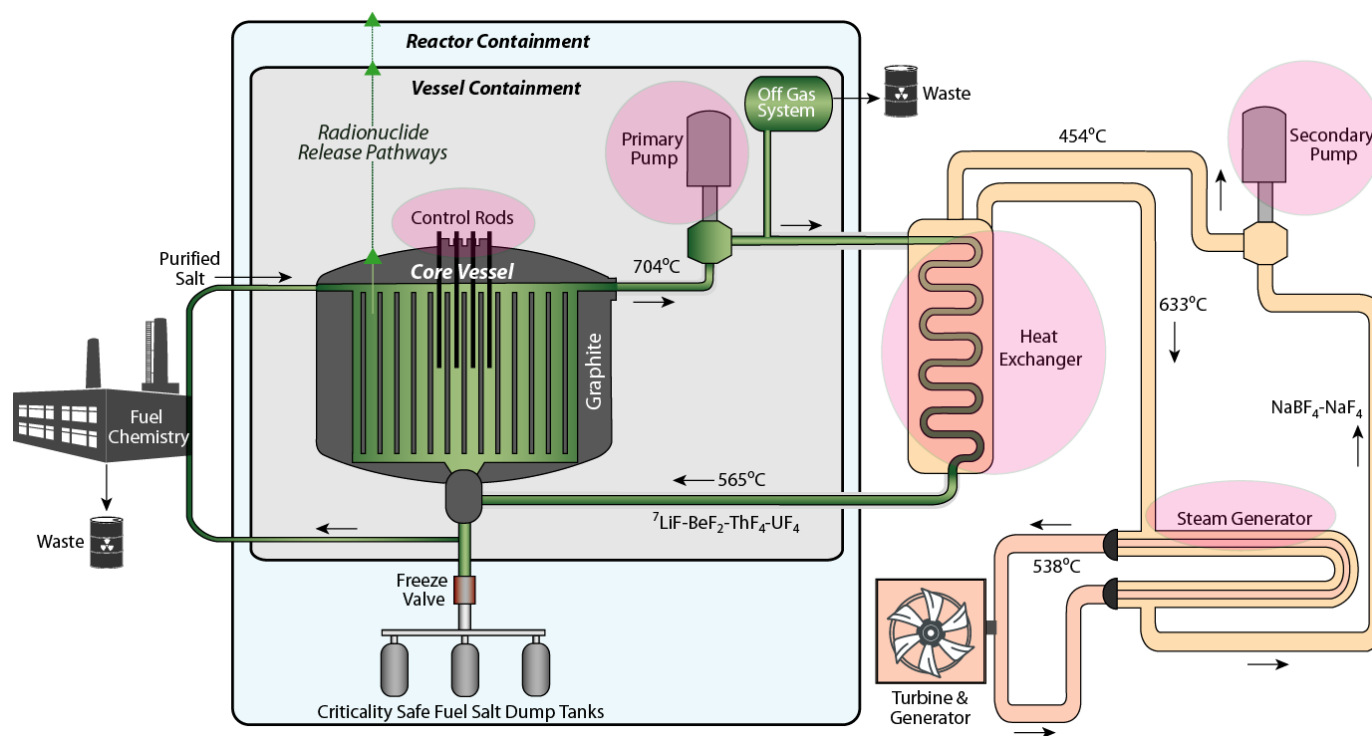


Figure 15. Schematic conceptual drawing of the MSBR concept considered in the accident analyses of Yoshioka et al. (2012). The reactor systems that could fail and possibly initiate transient scenarios are highlighted in red (adapted from Yoshioka et al., 2012).

The first step in an event chain that could lead to radionuclides being released to the environment from a MSBR would involve the rupture or leaking at the primary loop boundary, which consists of the reactor vessel, salt pipes, pumps, heat exchangers and other minor components. There are two primary ways in which a rupture of the primary loop could occur: (1) external cause accidents, and (2) internal cause accidents (Yoshioka et al., 2012).

The generic externally caused accidents include earthquakes, tsunamis, floods, wind, fire, human intrusion etc. The internally caused accidents involve over pressurization, overheating and other mechanical failures within the primary loop. Over pressurization and over temperature events can be caused by a power increase

accident or a flow decrease accident (temperature rises as power increases and temperature rises as flow decrease). Analyses of over-pressurization events indicate they are extremely unlikely for MSR due to the low vapor pressure of the fuel salts involved (e.g., Yoshioka et al., 2012). Even if the primary loop boundary is breached, there are multiple barriers to radionuclide release to the environment such as the reactor containment vessel and the reactor building (shown schematically in Figure 15). The following sections summarize several conceivable internally caused MSR accident scenarios that may need to be considered as part of a source term analysis.

3.1.1 *Power Increase (reactivity initiated) Transients*

For designs that include neutron absorbing control rods, a power increase transient can occur due to the accidental withdrawal or ejection of the rods due to equipment failure or operator error. For designs containing graphite control rods, the accidental insertion of the rods may cause a power increase transient. In either case the result would be an increase in temperature. It is not likely that such an event would result in a significant power excursion due to the negative reactivity feedback of fuel salt with increasing temperature. Power increase transient scenarios for a breeder type MSR have been analyzed in some detail by Shimazu (1978) and Suzuki and Shimazu (2008).

The negative reactivity feedback of the fuel-salt with temperature could lead to a power increase transient if cold-fuel-salt is accidentally injected into the core. This could occur, for example, if the fuel pump is inadvertently started from a stand-by-condition. However, due to the relatively small decrease in core temperature that would be anticipated for such an event, this type of transient has been deemed to be of limited consequence (e.g., Yoshioka et al., 2012). This type of increased reactivity could also be caused by the fuel-salt experiencing more cooling than normal operations call for. This could occur if there is an increase in the secondary coolant salt flow or if fuel-salt flow is accidentally increased, thus lowering core temperatures (Yoshioka et al., 2012).

Excess reactivity may also be added to the core through the addition of excess fissile material due to equipment failure or errors associated with the operation of the fuel-salt composition adjustment system. The same effect would be achieved if neutron absorbing materials such as thorium salts were removed from the system (Yoshioka et al., 2012).

De-pressurization of the fuel-salt could also lead to an increased power transient. This could occur because of the positive void reactivity coefficient of most fuel-salts and the presence of inert gas bubbles introduced to sparge gaseous fission products. A decrease in pressure in the fuel-salt could lead to the sparge gas bubbles becoming larger, thus initiating an increase in reactivity (Yoshioka et al., 2012). Such depressurization could be caused by a rupture in the primary loop or pump failure. This process could also be caused by problems with the inert gas bubble injection system (Yoshioka et al., 2012).

If uranium were to precipitate due to some unforeseen changes in fuel-salt chemistry and the solid uranium particles were injected into the core, a positive reactivity event would occur. Yoshioka et al., (2012) suggest that uranium precipitation could occur due to the exposure of the fuel salt to moisture and oxygen, presumably resulting in the conversion of uranium of uranyl oxides or oxyhydroxides.

3.1.2 *Flow Decrease Accidents*

An electrical failure of fuel pumps would lead to an increase fuel-salt temperature and an increase in delayed neutron flux within the core, which has the same effect as an increase in reactivity. As indicated by Yoshioka et al., (2012) this type of event would likely be of low consequence because of the negative reactivity feedback of the fuel-salt with increasing temperature; furthermore, control rods would be inserted as soon as the fuel pumps tripped.

A possibly more severe transient could result if the fuel pump actually seized up (immobilizing pump shaft) because the natural convective circulation of fuel-salt would be halted leading to a rapid temperature increase in the core (Yoshioka et al., 2012). For example, if for some reason control rods are not scrammed during a pump seizure event, Shimazu (1978) calculated that fuel salts in a MSBR could reach temperatures of around 900 °C approximately 300 seconds after all the fuel-salt pumps lock-up. If control rods did deploy, the consequence of a fuel pump seize-up event would be less severe but still result in a rapid core temperature excursion (Yoshioka et al., 2012).

The loss of cooling from the secondary-salt due to rupture or pump failure would also cause a rapid increase in core temperatures that, if rapid enough, could result in fuel-salt being drained into the criticality-safe dump tanks (see Figure 15). The result of a fuel-salt flow blockage within the primary loop would have similar consequences to the pump failure or seizure transients. However; it was deemed highly unlikely that the blockage of all channels would occur (Yoshioka et al., 2012).

If there is a rupture in the vessel, pipes, pumps or heat exchangers, fuel-salt may leak into a catch-pan or emergency drain tank. The dispersal of radionuclides during such an event will depend on the amount of fuel-salt lost and the integrity of other systems. In the event of a heat exchanger rupture, fuel-salt may mix with secondary coolant salt leading to a change in chemistry that could influence radionuclide mobility. Ruptures can occur due to thermal stress cycling, high temperature corrosion or mechanical failure due to manufacturing flaws (Yoshioka et al., 2012).

Other possible transients include the rupture of the steam generator which could cause damage to the secondary loop and must be isolated from the primary loop to avoid a severe accident consequence propagation (Yoshioka et al., 2012). Rupture or failure of pipes could also be caused during a re-melting operation due to the significant volume expansion of most fuel-salts when going from solid to liquid state. If a pipe or tank that is nearly completely filled with frozen salt is heated too rapidly, expansion may lead to enhanced stress corrosion or immediate failure (Yoshioka et al., 2012).

A fuel-salt flow block event could also occur due to salt freezing. This could conceivably happen if one of the fuel pumps failed leading to the over cooling of fuel salt adjacent to the heat exchanger (assuming that the heat exchanger inlet temperature is lower than the freezing point of the fuel) (Yoshioka et al., 2012). One possible failure that could lead to transfer of radionuclides from the core to the reactor containment volume would be a failure within the off-gas system. Such a failure would result in radioactive noble gases, tritium and any other radionuclides that have accumulated in the off-gas, such as iodine and cesium, being made available for transport within the containment vessel (Yoshioka et al., 2012).

3.2 Summary of the Safety Assessment of the Molten Salt Fast Reactor Project

Initial source term work on a European molten salt fast reactor (MSFR) concept is described in Gérardin et al., (2019). This source term project is being performed as part of the Safety Assessment of the Molten Salt Fast Reactor project (SAMOFAR) and involves two complementary conceptual approaches based on established risk assessment principals (discussed below). The stated goal of the SAMOFAR project is to demonstrate the innovative safety concepts of the MSFR through advanced experimental and numerical techniques (Kloosterman, 2017). The MSFR is based on the molten salt breeder reactor designed at Oak Ridge National Laboratory in the 1960s. The MSFR design being considered consists of a 2.25 meter diameter cylindrical reactor vessel made of a nickel-based alloy containing liquid fuel at ambient pressures at an operating temperature of 750 °C (Kloosterman, 2017). The reactor is designed with a fast neutron spectrum and can be operated as a breeder or burner due to its flexible fuel salt composition that can be adapted during reactor operation.

During operation of the MSFR, a fraction of the fuel salt is continuously diverted to a salt clean-up unit located outside of the core to extract lanthanides and actinides (Kloosterman, 2017). The salt clean-up process involves two steps (Kloosterman, 2017):

- Gaseous and insoluble fission products (e.g., noble metals) are removed from the primary circuit by gas bubbling near the primary fuel salt pumps.
- Uranium, actinides, and some fission products are separated by using pyrochemical techniques (fluorination and reductive extraction) in a chemical plant located outside of the core region.

Both of these systems should be considered as part of an MSR source term analyses.

The key safety features of an ambient pressure, liquid fueled salt reactor (such as the MSFR) that will play important roles in an MSR source term analysis include (Kloosterman, 2017):

- Thermal expansion of the fuel salt. Reactivity excursions are mitigated by the expansion of the fuel salt upon heating, which gives a strong negative void and temperature reactivity feedback everywhere in the core. The resulting temperature feedback coefficient is around 25 pcm/K.
- Natural circulation of the fuel salt. As long as the primary circuit is not blocked, decay heat is removed from the fuel salt by circulation through the heat exchangers even if the fuel pumps are not operating. This safety feature is enhanced by the fact that the fuel salt has a high heat capacity and the system is operated well below its boiling point.
- Draining of the fuel salt. In the event of a significant off-normal situation, the fuel drains into passively cooled dump tanks (with subcritical geometries) located under the core. The core vessel is connected to the drain tanks by a freeze plug that melts if the fuel salt is overheated (e.g., if external power fails).
- Cleaning the fuel salt. The effect of decay heat is also minimized by removing fission products from the fuel salt continuously by helium bubbling (sweep gas) and chemical separations (see bullets above). The cleaning process has been shown to reduce decay heat in the salt by around 35%.
- High solubility of fission products in the salt. Fission products that are volatile in other types of reactors (e.g., cesium and iodine) are retained in the fuel salt, thereby potentially reducing reactor core source term.

The objective of the SAMOFAR project is to validate and quantify each of these safety features through advanced experimental studies and modeling and simulation tools.

3.2.1 Methodology of Transient Scenario Identification

Source term analysis of the MSFR concept in the SAMOFAR project uses two complementary but conceptually distinct approaches to identify potential accident initiating events (Gérardin et al., 2019). Because these safety analyses methodologies were developed while reactor design and operating procedures (start up, shutdown, maintenance) were still being defined, the approaches needed to include methods that could be applied without knowledge of specific components, systems and procedures (Gérardin et al., 2019). The two approaches chosen to provide maximum flexibility were the Master Logic Diagram (MLD) top down approach and the Functional Failure Mode and Effect Analyses (FFMEA) bottom-up approach. In the FFMEA approach, the user is led to reason from a functional point of view by looking for the possible causes of the loss of functions and the resulting consequences, whereas the MLD approach is more focused on phenomena and involves identifying the causes of processes that may lead to the degradation of radionuclide barriers such as the core vessel (Gérardin et al., 2019).

For the MLD approach, risks have been differentiated according to the physical phenomena involved. This method also reveals risks caused by external hazards or by failures in other systems of the plant. The FFMEA approach reveals more about failure modes because it links the loss of each function to a generic component or system failure (Gérardin et al., 2019). Therefore, the FFMEA approach has the advantage of providing information on the systems and procedures used for detection, prevention and mitigation of events. The MLD is a good graphical tool for presenting possible hazards and elucidates the logical connections between hazards.

The FFMEA approach is a qualitative methodology used to define possible incident and accident initiators when sufficient design detail is not available to support evaluations at the component level. The FFMEA table is compiled by postulating the loss of functions rather than specifying specific component and systems failures (Gérardin et al., 2019). An example of a small section of a FFMEA table is shown in Table 11.

Table 11. Example section of a FFMEA table (adapted from Gérardin et al., 2019).

Reactor Section	Process Function	Failure Mode	Cause of Failure	Consequences of Failure	Potential Transient Initiating Event
Core vessel	Contain fuel-salt and fission products.	Loss of containment	Leak/Rupture	<ul style="list-style-type: none"> Fuel-salt flows outside of core. Possible shutdown of criticality if enough fissile material is lost from core. Collection of fuel in subcritical catch basin. 	Stress and/or corrosion caused cracking.

The MLD approach is a qualitative risk analysis method that identifies hazards and possible initiating events of a system through a structured deductive approach. It is a “top-down” method that is well suited for scenarios where component and systems designs have not been finalized as the analysis is based on possible physical phenomena rather than on specific components of a design. The MLD tool also highlights the correlations between different functions or phenomena and can, therefore, be an asset in the study of complex systems such as an MSR (Gérardin et al., 2019).

The general steps of the MLD approach are to (Gérardin et al., 2019):

- Identify main undesired transient event to be prevented.
- Decompose main transient event into detailed sub-events that must occur for transient to take place.
- Continue decomposition until all physically possible phenomena and associated events that directly challenge safety functions are identified.
- Identify initiating events, which are the basic phenomena that cannot be further divided in to sub-events.

The conceptually distinct hazard FFMEA and MLD analyses methods provide robust tools for identifying hazards and initiating events for the MSFR concept. These tools helped highlight the open points in the concept (undefined components and systems) that could represent design opportunities to mitigate some of the identified hazards (Gérardin et al., 2019). Table 12 provides an excerpt from the MSFR source term study showing key postulated accident initiating events identified using the FFMEA and MLD tools (Gérardin et al., 2019).

Table 12. Excerpt from the MSFR source term study showing key postulated accident initiating events identified using the FFMEA and MLD tools (adapted from Gérardin et al., 2019)

Loss of Flow/Cooling	<ul style="list-style-type: none"> • Complete loss of intermediate salt • Fuel salt freezing scenario • Blockage of all sectors of fuel salt circuit • Complete rupture or blockage of all the fuel circuit pumps • Overworking of all the fuel circuit pumps • Conversion circuit pumps overworking • Over-working of the pumps of the intermediate circuit • Obstruction/blockage of the intermediate circuit • Rupture/blockage of all intermediate circuit pumps • Obstruction of all free levels (including the vertical inlet pipe from the core to the expansion vessel) • Total loss of electric power
Structural Degradation Breach/Rupture	<ul style="list-style-type: none"> • Important deformation of the fuel circuit possibly leading to an increased core volume (e.g. the welded joints taking the recirculation sectors in the correct position collapse) • Breach in the core vessel • Breach in the lower reflector (with rupture of the structure cooling system) • Breach in the upper reflector with rupture of the structure cooling system and/or with damages to the expansion vessel system • Breach of a heat exchanger plate/channel • Rupture of blanket tank wall between fuel and fertile salt with rupture of the fuel circuit walls cooling circuit • Rupture of a pipe of the reactivity control system • Rupture of the connection between the free surface of the fuel storage tank and the free surface of the core for the gas in the part between the core cavity and the valve • Detachment of the wall thermal protection • Complete rupture of the pressurized sampling device
Degradation of Off-gas system	<ul style="list-style-type: none"> • Rupture/obstruction of reactivity bubble injector • Rupture of horizontal bubble injector for salt cleaning • Rupture of the gas separation chamber • Rupture of the gas processing unit (with possible leak of processing fluid)
Off-normal Chemistry	<ul style="list-style-type: none"> • Bulk precipitation of fissile matter (e.g. because of an inlet of water) • Undetected deviation of the chemical composition • Chemical reaction between different fluids (e.g. hot part of intermediate circuit, water)
Reactivity Insertion	<ul style="list-style-type: none"> • Accidental insertion of fuel • Prompt critical power excursion with induced shockwave

A list of the postulated accident initiating events For the MSFR source term analyses is shown in Table 12. This can be used to define reference accident scenarios that can be deterministically modeled (mechanistic source term) to assess the severity of the phenomena, transient behavior and consequences of a particular scenario (Gérardin et al., 2019). The list of postulated initiating events will evolve as the design is refined and the deterministic analysis of components and transients are performed. Some initiating events will be removed from the list based on mitigating systems added to the design (i.e., systems that make the event physically impossible) (Gérardin et al., 2019). Each accident scenario is classified into frequency and consequence categories, where the consequences are ranked based on severity of damage to the reactor and extent of environmental release. Risk matrices are built using the accident scenario categorization information (Gérardin et al., 2019).

4.0 Recommendations and Future Work

In addition to the significant progress made in accident analyses for some MSR concepts (see examples discussed in Section 3.0 above), there remains a need for a thorough, step-by-step source term analyses that includes parametric sensitivity analyses and trial calculations. The SFR mechanistic source term study presented in Grabaskas et al., [2016a] and summarized in Section 2.0 above, represents an excellent template on which to base future MSR source term work.

It is recommended that the MSR source term work take the parallel-path approach used for the SFR project through the development of two types of models:

1. A simplified mass balance model that be compiled from existing information and tools that can be used for sensitivity studies to rank the importance of specific radioisotopes and release pathways in term of off-site dose consequence.
2. A mechanistic model that provides a realistic, science-based assessment of the radionuclide release and retention that accounts for all key transport phenomena. This mechanistic MSR source term model would provide feedback to reactor design teams regarding the impact of design features and could eventually provide a means of justification for reducing the size of emergency planning zones and site boundaries for new MSR reactors.

The risk analyses tools discussed in Section 3.2 above [i.e., the master logic diagram (MLD) and functional failure mode effect analyses (FFMEA)] will be used in the determination of potential accident initiating events and will guide the selection of transient scenarios. These tools are particularly applicable to reactor concepts that are still in the design phase, such as MSRs, because they focus on evaluating fundamental radionuclide release phenomena and the loss of safety system functionality rather than the failure of specific reactor components and systems. This allows the reference reactor for the source term model to remain largely generic in the sense that the design contains open-points for which specific components and mechanisms have not yet been specified. Such an approach allows the source term model to provide feedback into the reactor design process.

Another important guide that will be used in the MSR source term study is the ASME/ANS, 2013 standard “Probabilistic Risk Assessment Standard for Advanced Non-LWR Nuclear Power Plants” that is summarized in Section 1.0 above. This standard proposes a sound methodology for performing risk assessments of advanced, non-LWR nuclear power plants and provides guidance and requirements for the source term analyses. The source term guidance is technology neutral and compatible with the FFMEA and MLD risk analyses tools discussed above.

The proposed path forward for the MSR source term project is shown in Figure 17. This project would involve collaborative parallel activities involving reactor engineers, risk analysts, chemists and computational scientists and would thus provide a rigorous and generically applicable MSR source term analyses tool.

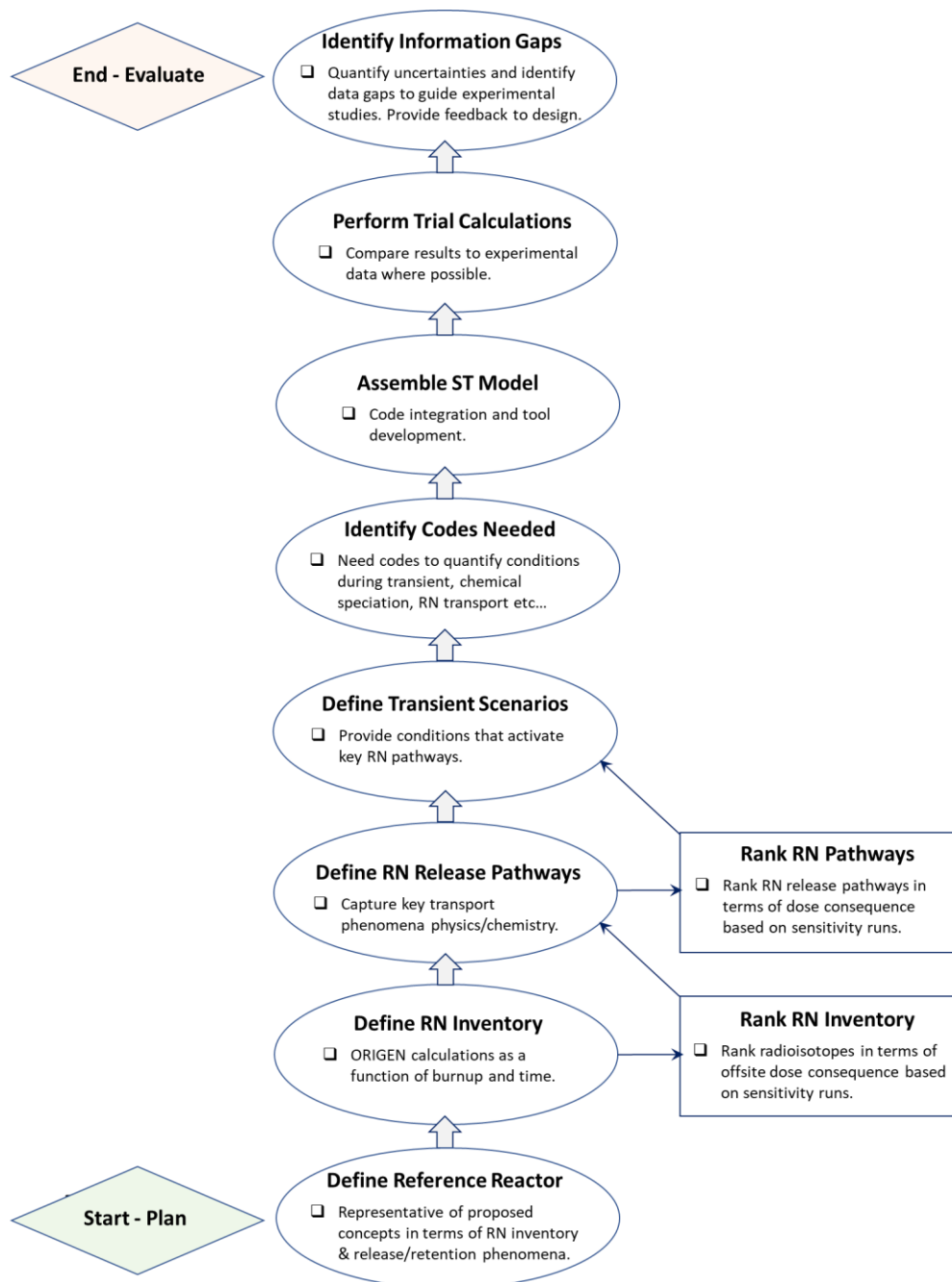


Figure 17. Conceptual pathway for developing a rigorous source term analysis for MSRs.

References

ASME/ANS, "Probabilistic Risk Assessment Standard for Advanced Non-LWR Nuclear Power Plants," Joint Committee on Nuclear Risk Management standard RA-S-1.4-2013, 2013.

Beall, S. E., Haubenreich, P. N., Lindauer, R. B., Tallackson, J. R., "MSRE Design and Operations Report Part-V, Reactor Safety Analysis Report", ORNL-TM-732, 1964.

Gérardin, D., Ugenti, C. A., Beils, S., Carpignano, A., Dulla, S., Merle, E., Heuer, D., Laureau, A., Allibert, M., "A methodology for the identification of the postulated initiating events of the Molten Salt Fast Reactor", *Nuclear Engineering and Technology* 51, 1024 -1031, 2019.

Grabaskas, D., Brunett, A. J., M. Bucknor, M., J. Sienicki, J., Sofu, T., 2015, "Regulatory Technology Development Plan - Sodium Fast Reactor: Mechanistic Source Term Development," Argonne National Laboratory ANL-ART-3, 2015.

Grabaskas, D., Bucknor, M., Jerden, J., Brunett, A. J., Denman, M., Clark, A., Denning, R. S., "Regulatory Technology Development Plan-Sodium Fast Reactor: Mechanistic Source Term–Trial Calculation", ANL-ART-49, Argonne National Laboratory, 2016a.

Grabaskas, D., Bucknor, M., Jerden, J., "Regulatory Technology Development Plan - Sodium Fast Reactor: Mechanistic Source Term Development - Metal Fuel Radionuclide Release," ANL-ART-38, 2016b.

Haubenreich, P. N., Engel, J. R., 1962, "Safety Calculations for MSRE", ORNL-TM-251, 1962

International Atomic Energy Agency (IAEA), "Fission and Corrosion Product Behaviour in Liquid Metal Fast Breeder Reactors (LMFBRs)," IAEA-TECDOC-687, 1993.

Jerden, J. Grabaskas, D., Bucknor, M., "Development of a Thermochemical Database for Sodium Fast Reactor Mechanistic Source Term Calculations", proceedings paper, ANS Annual Meeting, June 9 – 13, 2019.

Kloosterman, J. L., "Safety assessment of the molten salt fast reactor (SAMOFAR)", in *Molten Salt Reactors and Thorium Energy*, pages 565-570, 2017.

McFarlane J., Weber, C. F., Greenwood, M. S., Qualls, A. L., "Thermochemical and Transport Properties Important to Molten Salt Reactor Operation: Off-gas Performance and the Fission Product Mechanistic Source Term", ORNL/TM-2018/958, 2018.

Osterhout, M., "Control of Oxygen, Hydrogen, and Tritium in Sodium Systems at Experimental Breeder Reactor II," UAC-41069, 1978.

Outotec, HSC Chemistry 8 User's Guide, 2014.

Shimazu Y., "Nuclear Safety Analysis of a Molten Salt Breeder Reactor", *Journal of Nuclear and Science Technology*, Vol.15, No. 7, 1978.

Shimazu, Y., "Locked Rotor Accident Analysis in a Molten Salt Breeder Reactor", *Journal of Nuclear Science and Technology*, Vol. 15, No. 12, 1978.

Suzuki, N., Shimazu, Y., "Reactivity-Initiated-Accident Analysis without Scram of a Molten Salt Reactor", *Journal of Nuclear and Science Technology*, Vol. 45, No.6, 2008.

Thomas, J. W., "Validation of the Integration of CFD and SAS4A/SASSYS-1: Analysis of EBR-II Shutdown Heat Removal Test 17", in Proceedings of the *International Congress on Advances in Nuclear Power Plants (ICAPP 2012)*, Chicago, IL, 2012.

NRC, "Issues Pertaining to the Advanced Reactor (PRISM, MHTGR, and PIUS) and CANDU 3 Designs and their Relationship to Current Regulatory Requirements," U.S. Nuclear Regulatory Commission SECY-93-092, 1993.

NRC, Accident Source Terms for Light-Water Nuclear Power Plants, U.S. Nuclear Regulatory Commission report NUREG-1465, 1995.

NRC, "Policy Issues Related to Licensing Non-Light Water Reactor Designs," U.S. Nuclear Regulatory Commission report SECY-03-0047, 2003.

NRC, "Second Status Paper on the Staff's Proposed Regulatory Structure for New Plant Licensing and Update on Policy Issues Related to New Plant Licensing," U.S. Nuclear Regulatory Commission report SECY-05-0006, 2005.

Yoshioka, R., Shimazu, Y., Mitachi, K., "Guidelines for MSR Accident Analysis", *Thorium Energy Conference (ThEC12)*, Shanghai, October 2012.



Chemical & Fuel Cycle Technologies Division

Argonne National Laboratory
9700 South Cass Avenue, Bldg. 208
Argonne, IL 60439-4842

www.anl.gov



Argonne National Laboratory is a U.S. Department of Energy
laboratory managed by UChicago Argonne, LLC